# <sup>137</sup>Cs AND <sup>210</sup>Pb TRANSPORT AND GEOCHRONOLOGIES IN URBANIZED RESERVOIRS WITH RAPIDLY INCREASING SEDIMENTATION RATES

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(Accepted for publication November 30, 1983)

#### ABSTRACT

McCall, P.L., Robbins, J.A. and Matisoff, G., 1984. <sup>137</sup>Cs and <sup>210</sup>Pb transport and geochronologies in urbanized reservoirs with rapidly increasing sedimentation rates. In: J.A. Robbins (Guest-Editor), Geochronology of Recent Deposits. Chem. Geol., 44: 33-65.

Sedimentation rates have been measured in three reservoirs in northeastern Ohio, U.S.A., by means of 137Cs and 210Pb geochronologies, volumetric surveys and varve counting. These various methods, while only partially overlapping for each reservoir, show dramatic (three-fold) increases in rates of sediment accumulation in each system between about 1940 and 1977. Mass sedimentation rates are very nearly proportional to the size of the population in the region and possess a doubling time of roughly 19 yr. In these systems with changing sedimentation rates, the preferred model for use with 210Pb geochronologies is one which assumes a constant activity of material added to surface sediments. In systems possessing large (watershed)/(reservoir area) ratios, increasing erosion is evidently accompanied by a proportionate increase in the erosion of excess 210Pb. High near-surface activities of 137Cs are due to system integration effects with time constants the order of 10 yr. to a few decades. Total accumulation of fallout 137Cs and excess 210Pb far exceed direct atmospheric loadings, thus indicating the importance of watershed contributions and implying annual retention of the radionuclides in the reservoirs of between ~15% and ~80%. In Lake Rockwell, sedimentary fluxes of Zn, Pb and Cu have increased with time. The flux of Cu in particular has increased very markedly since 1970, and concentrations are high in surface materials due in large part to addition of CuSO<sub>4</sub>, an algicide, to the water. Because of increased rates of sedimentation, the remaining useful life of Lake Rockwell has decreased from 203 to 67 yr., while the remaining useful life of Mayfair Lake is now less than 5 yr.

# INTRODUCTION

Multiple geochronological methods, including radionuclide chronologies, have seldom been applied to studies of sedimentation in reservoirs. Because

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these man-made systems are sensitive to land-use changes during the past several decades, they provide a means for comparing and contrasting alternative geochronological methods and models. In this paper, net rates of sediment accumulation are determined in cores from three freshwater reservoirs in northern Ohio, U.S.A., by a partially overlapping combination of methods including radiometric age dating (via <sup>137</sup>Cs and <sup>210</sup>Pb), volumetric sediment surveys, and varve stratigraphy. Sedimentation rate data are of value because the accumulation of sediments shortens the useful life of reservoirs and landuse-related changes in sediment accumulation may indicate a shorter reservoir lifetime than may have been previously anticipated. Apart from accumulation effects, sediments constitute a major contaminant in sources of drinking water. Increased urbanization of the drainage basin of these reservoirs has resulted in increased contaminant loads which may be chronicled in accumulating sediments. Sedimentary records of metal and organic contaminants can in principle be used to reconstruct exposures of populations using a particular water supply. Since these reservoirs were created at known times, core dating techniques may be usefully compared with volumetric surveys and inferences made concerning non-steady-state transport characteristics of these systems.

#### **METHODS**

Two of the reservoirs studied are located on the Cuyahoga River or its tributaries. The Cuyahoga River has its headwaters in northeastern Ohio and runs south to the city of Akron, Ohio, where it turns north and enters Lake Erie at Cleveland, Ohio. The reservoir closest to the headwaters of the Cuyahoga is East Branch Reservoir, created in 1939 on East Branch River (Fig. 1). East Branch Reservoir has a surface area of 1.70 km<sup>2</sup> and a drainage basin area of 45.3 km<sup>2</sup> (Hahn, 1955). It is used for recreation and river-level control. Lake Rockwell is an impoundment of the Cuyahoga River itself, and was made in 1914 for the sole use as a water supply for the city of Akron, Ohio. It occupies a surface area of 2.78 km<sup>2</sup> and has a drainage basin area of 531 km<sup>2</sup> (Hahn, 1955). Mayfair Lake is located in Richmond Heights, Ohio, on a tributary to Euclid Creek and was made some time before 1942 (approximately 1938) for recreational use. It occupies an area of 0.016 km<sup>2</sup> and has a drainage basin area of 2.4 km<sup>2</sup>. Surface deposits in most of the drainage basin of the Cuyahoga River consist of Quaternary glacial sands, silts and clays; in a few places below Lake Rockwell the river has cut through glacial deposits into bedrock which consists of Paleozoic conglomerates, sandstones and shales (Hough, 1958). The surface sediments in the reservoirs have water contents of 70–80%, are >90% silt—clay, and contain 9–11% organic matter by weight.

In 1977, two 6.5-cm diameter cores from Lake Rockwell and one from East Branch Reservoir were collected by divers for radiometric analysis (Fig. 2). An additional core was collected from Mayfair Lake in 1982. The

cores were extruded in the laboratory and cut into 1-cm sections to a depth of 24 cm, then into 2-cm sections to a depth of 40 cm, and finally into 5-cm sections for the remaining length of the cores. Core sections were weighed, air-dried and reweighed to calculate sediment bulk density as a function of depth in the core, and ground and passed through a 60-mesh sieve.

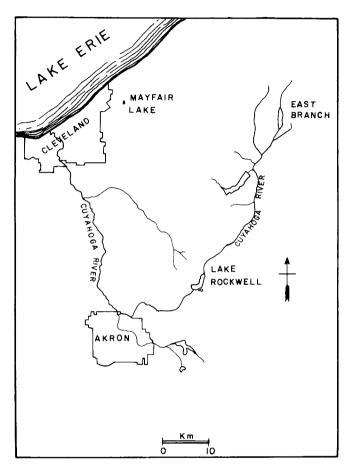


Fig. 1. Location of the three reservoirs in northeast Ohio.

The activity of  $^{137}\mathrm{Cs}$  in core sections was determined with a  $2\times2$  in.  $(5.08\times5.08~\mathrm{cm})$  NaI detector coupled to a multichannel analyzer—counter—timer system. Counting times ranged from 2 to 15 hr. per sample. As  $^{137}\mathrm{Cs}$  is a high-energy gamma emitter (0.661 MeV), sediment self-absorption effects are practically eliminated (Robbins et al., 1978). Activities were corrected for sediment porosity and expressed as activity per gram dry weight of sediment. For determination of the activity of  $^{210}\mathrm{Pb}$ , 3-g portions of dry sediment were extracted in a mixture of hot (86°C) 50% HCl

for a period of 36 hr. Periodically 30%  $\rm H_2O_2$  was added during the extraction period to facilitate destruction of the organic matter. The decay product of  $^{210}$  Pb,  $^{210}$ Po, was plated from pH-adjusted extracts onto polished silver discs and counted on a gas flow proportional counter. Overall chemical yield and plating efficiencies were determined by sequential plating of the acid extracts and were generally better than 90%. Values of the  $^{210}$ Pb activity are expressed in pCi g $^{-1}$ .

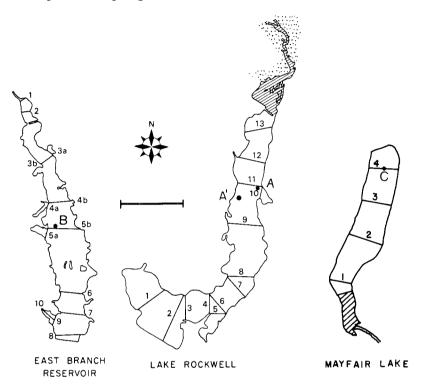


Fig. 2. The three reservoirs studied. (Bar length = 1 km for East Branch Reservoir and for Lake Rockwell; = 0.12 km for Mayfair Lake.) The *numbered lines* indicate cross-reservoir transects for volumetric surveys. Cores for radionuclide lettered sites. In Lake Rockwell A = core 1 and A' = core 2. The core for varve analysis was collected at site C in Mayfair Lake.

Volumetric surveys of the sediment in Lake Rockwell and East Branch were conducted in 1977. Transects were laid out perpendicular to the long axis of each reservoir as shown in Fig. 2. At seven equidistant points along each transect a ruled plumb line affixed to a 0.04-m<sup>2</sup> flat plate weighing 2 lb. (0.907 kg) was used to measure the depth of the water overlying the sediment—water interface, and a ruled length of  $\frac{3}{4}$ -in. (1.9 cm) galvanized pipe was used to measure the thickness of sediment over the original basin floor. Where the original reservoir floor consisted of sand- and gravel-sized

glacial till, the thickness of the predominantly silt—clay-sized sediments deposited since the creation of the reservoir was accurately determined. Where the original floor consisted of vegetated soil (determined in sediment cores by the presence of plant roots), poles often penetrated the water-logged soil profile and gave an overestimate of sediment thickness. The addition of 3-in. (7.62 cm) diameter floor flanges to the bottom of the poles for the Lake Rockwell survey corrected this problem. The same technique was used in Mayfair Lake, except that only three measurements were made along each transect (Fig. 2). Knowing the elevation of the water surface, sediment water interface, and sediment thickness, we calculated the cross-sectional area of water and sediment along each transect and graphically integrated these areas along the axis of the reservoir to obtain water and sediment volumes using the methods of Heineman and Dyorak (1963). Volume of sediment in major inlets was calculated from transect data using the modified Eakins range end formula (Gottschalk, 1951). These two volumes were then summed to obtain an estimate of the volume of sediment in the reservoir.

Sediment cores were collected from various parts of the reservoirs to check the accuracy of the poling method and to look for internal sedimentary structures and evidence of trace-metal pollution that often accompanies increased human use of a watershed. Internal sedimentary structures were examined in radiographs of the cores. Sediment metal concentrations of Zn, Cu, Pb and Fe were determined by analysis of the acid-extractable portion of bulk sediments using atomic absorption spectrophotometry (A.P.H.A., 1976; Perkin-Elmer, Inc., 1974).

## RESULTS AND DISCUSSION

## Internal sedimentary structures

Radiographs of a sediment core from Lake Rockwell exhibited few internal sedimentary structures (varves), despite the fact that macrobenthos abundances in the core were low (166 chironomids/m², 200 tubificids/m²). This is probably due to the fact that most river-borne coarse-grained sediments are trapped in the relatively flat region upstream of the lake. The seasonal variation in river flow thus does not produce a seasonal variation in the type of sediment delivered to the lake, so only fine-grained and relatively low-density material is present within the basin.

Mayfair Lake is considerably smaller than Lake Rockwell; there is no extensive flat-lying area to trap sediments above the lake. The core collected from Mayfair Lake shows extensive sediment banding (Fig. 3). Mayfair Lake is  $\sim 40$  yr. old. There are 52 discernible pairs of low- and high-density sediment bands in the core; but some of these are subcouplets that probably represent flood deposition within seasonal bands. Based on the size of surrounding bands, there are 44 couplets which are annual bands with three

distinct zones visible. The first extends from the sediment—water interface to 12 cm; 5 couplets can be detected in the radiographs, yielding a sedimentation rate from 1977 to 1982 of 2.4 cm yr.<sup>-1</sup>. The second zone encompasses approximately the period 1956—1977 where the sedimentation rate is 1.29 cm yr.<sup>-1</sup>. The lowest zone extends from 1956 to reservoir closure (approximately 1938) where the sedimentation rate is 0.83 cm yr.<sup>-1</sup>.

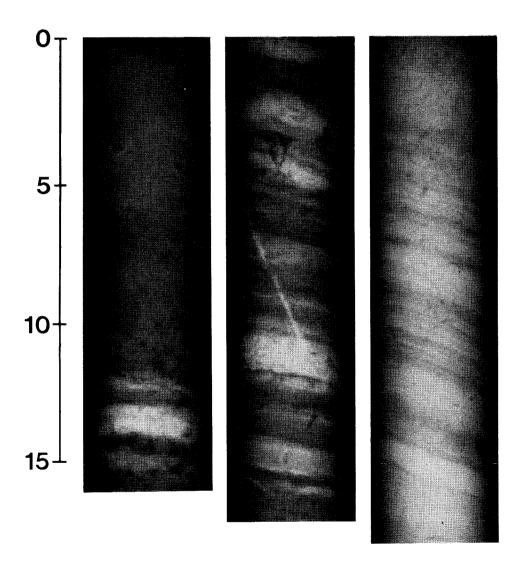


Fig. 3. Stratigraphy of the Mayfair Lake core. The topmost section of the core is on the left. Dark bands indicate sediments of relatively low density.

## Volumetric surveys

The cross-sectional areas of sediment and overlying water along transects across the three reservoirs measured by poling are shown in Fig. 4. Lakes Rockwell and Mayfair are filling from the point of entry of the river into their basin. Marshes exist at the north end of Lake Rockwell which are not present on the 1944 topographic maps. The delta tops in Rockwell and Mayfair are shown as shaded areas in Fig. 2. Sediments are distributed fairly evenly along the remainder of each basin. Sediment cores taken in most parts of the reservoirs ought to be representative of the entire basin (the position of the sediment cores described above are shown in Fig. 2). The average annual volume of sediment accumulating in a reservoir was calculated by measuring the area under the sediment cross-sectional curves in Fig. 4 and dividing by the age of the reservoirs. Dividing again by the surface area occupied by the basin yields a long-term average basin-wide sedimentation rate (Table I).

TABLE I

Characteristics of three reservoirs and their watersheds

	Lake Rockwell	East Branch Reservoir	Mayfair Lake
Area of drainage basin (10 <sup>6</sup> m <sup>2</sup> )	531	45.3	2.4
Date of reservoir construction	1914	1939	1938 (estimated)
Date of this survey	1977	1977	1982
Basin volume (10 <sup>6</sup> m <sup>3</sup> )	7.42	5.57	0.018
Water volume (106 m³)	6.2	5.03	0.00892
Sediment volume (10 <sup>5</sup> m <sup>3</sup> )	8.22	5.4	0.0912
Average annual volume of sediment			
accumulated since closure (10 <sup>4</sup> m <sup>3</sup> yr. <sup>-1</sup> )	1.3	1.42	0.0228
Reservoir surface area (10 <sup>6</sup> m <sup>2</sup> )	2.78	1.70	0.0155
Area ratio:			
(drainage basin)/(reservoir)	191	27	155
Mean annual discharge rate (10 <sup>5</sup> m <sup>3</sup> day <sup>-1</sup> )	6.9		
Hydraulic residence time (days)	9.0		_

Sedimentation rates in Lake Rockwell and East Branch reservoirs have been examined by volumetric surveys in previous years (Tables I and IV). While East Branch Reservoir gives little indication of an increase in sedimentation rate between this survey and that in 1950, the Lake Rockwell surveys clearly show an increasing rate of sedimentation since 1950. The surveys of Hahn (1955) done in 1950 may have overestimated sediment thickness, since there is no indication that pole flanges were used to correct for penetration into a water-logged soil profile. However, in the analysis

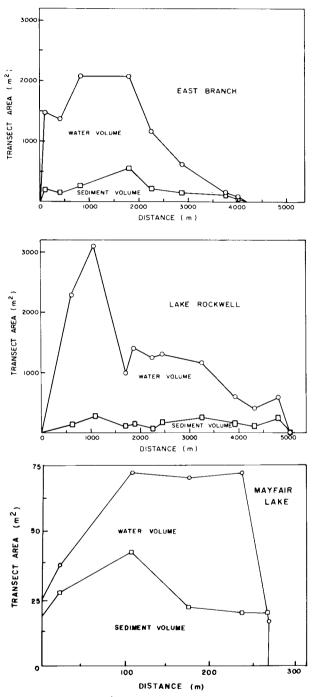


Fig. 4. Mean cross-sectional areas of water and sediment along numbered transects vs. longitudinal distance along the reservoir axes. The area under the bottom curve is the sediment volume.

below, the rate inferred by Hahn appears to be more consistent with the radionuclide data and population model (see Table IV).

# Profiles of 137Cs

This radionuclide  $(t_{1/2} = 30.2 \text{ yr.})$  is one of several long-lived isotopes produced by the atmospheric testing of nuclear weapons. As there is no natural occurrence of <sup>137</sup>Cs, the isotope was not present in the environment until the early 1950's. Maximum fallout to water and land occurred during 1963 and 1964 corresponding to the period of maximum testing activity by the U.S.A. and U.S.S.R. In many soil—water systems <sup>137</sup>Cs is strongly (and largely irreversibly) sorbed on clay minerals (Tamura and Jacobs, 1960; Schulz, 1965) and therefore traces the movement and accumulation of particles with which they associate. Its accumulation in sediments has provided a means of inferring accumulation rates in a variety of freshwater environments (Ritchie et al., 1973; Pennington et al., 1976; Robbins and Edgington, 1975; Robbins et al., 1978). Most often the position of the horizon (corresponding to the first appearance of <sup>137</sup>Cs in the environment) or the peak (corresponding to the period of maximum deposition) has been used to determine sedimentation rates. In this paper we shall use the entire

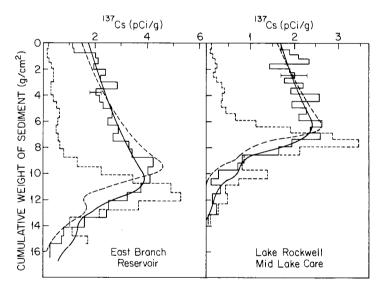


Fig. 5. Distribution of <sup>137</sup>Cs in the East Branch Reservoir and Lake Rockwell core 2. The solid histogram-like curves are actual values. The dashed line "histogram" is the distribution expected from the direct atmospheric fallout assuming a sedimentation rate which minimizes the discrepancy between measurement and this distribution. The solid smooth curve is the distribution expected if inputs are integrated prior to incorporation into sediments. The dashed curve is the distribution expected with integration and variable sedimentation rates based on the CA/population model (see text).

profile and examine the correspondence between the sedimentary record and the well-known loading of the isotope from the atmosphere.

The distribution of <sup>137</sup>Cs in two cores is shown in Fig. 5 (East Branch Reservoir and Lake Rockwell, core 2). The data are provided in Table II. A third profile (Lake Rockwell core 1) is shown in Fig. 6 along with excess lead <sup>210</sup>Pb discussed below. The data for this core are given in Table III. The solid histogram-like curve in the figure shows the measured distribution. The error bar attached to each curve provides an indication of the characteristic experimental uncertainties in the values. The histogram shown as the dashed line is that expected if the profile were the result of direct transfer of <sup>137</sup>Cs

TABLE II

Distribution of <sup>137</sup>Cs in cores from East Branch Reservoir and Lake Rockwell

Sediment	East Branch	Reservoir	Lake Rockw	ell (mid)	
interval (cm)	cumulative weight (g cm <sup>-2</sup> )	activity* (pCi g <sup>-1</sup> )	cumulative weight (g cm <sup>-2</sup> )	activity* (pCi g <sup>-1</sup> )	
0-1	0.38	1.12	0.45	1.68	
1-2	0.73	1.15	0.80	1.88	
2-3	1.15	2.02	1.17	2.09	
3-4	1.53	2.13	1.52	2.34	
4-5	1.96	1.94	1.87	1.44	
56	2.44	2.40	2.20	1.89	
6-7	2.98	2.20	2.57	2.02	
7-8	3.50	2.83	2.92	1.78	
89	4.02	2.01	3.40	2.23	
9-10	4.56	2.15	3.84	1.91	
10-11	5.16	2.28	4.39	2.57	
11-12	5.76	2.65	4.88	1.94	
12-13	6.33	2.50	5.36	2.27	
13-14	6.94	2.90	5.88	2.10	
14-15	7.52	2.69	6.35	2.64	
15-16	8.08	3.28	6.80	2.36	
16-17	8.75	3.80	7.31	2.61	
17-18	9.42	4.22	7.86	1.95	
18-19	10.09	4.02	8.52	1.66	
19-20	10.72	4.11	9.16	0.81	
20-21	11.45	3.77	9.68	0.71	
21 - 22	12.14	3.25	10.35	0.30	
22 - 23	12.75	2.53	10.86	0.11	
23 - 24	13.39	1.99	11.56	0.33	
24-26	14.17	1.60	12.30	0.15	
26 - 28	14.79	0.78	13.12	0.23	
28-30	15.40	0.76	13.91	0.07	
30-32	16.67	0.30	_	_	

<sup>\*</sup>Analytical uncertainties are generally around 10%.

from the atmosphere. (The choice of time scale for this profile is discussed below.) The atmospheric deposition curve is obtained from data published by the U.S. Health and Safety Laboratory (H.A.S.L., 1976) for the North American Great Lakes region. It can be seen that in all cores the peak position and horizon occur at sediment depths which are consistent with the choice of a single sedimentation rate for each core. However, there is far more activity in sediments deposited after about 1964 than can be

TABLE III Distribution of  $^{137}$ Cs and excess  $^{210}$ Pb in core No. I from Lake Rockwell

Sediment interval (cm)	Cumulative weight (g cm <sup>-2</sup> )	<sup>137</sup> Cs (pCi g <sup>-1</sup> )	<sup>210</sup> Pb <sub>exc</sub> (*)	Error (pCi g <sup>-1</sup> )	
0-1	0.25	1.38	20.23	1.76	
1-2	0.58	1.35	24.29	1.93	
23	0.95	1.61	23.17	1.81	
3-4	1.33	1.60	20.49	1.77	
4-5	1.77	1.84	21.57	1.78	
56	2.16	2.29	21.54	1.78	
6-7	2.53	1.63	22.02	1.79	
7-8	2.94	1.78	19.48	1.75	
8-9	3.36	1.60	19.99	1.76	
9-10	3.71	2.61	18.71	1.74	
10-11	4.09	2.40	18.90	1.74	
11-12	4.46	2.90	17.09	1.71	
12-13	4.79	2.17	20.97	1.77	
13-14	5.11	2.17	21.22	1.73	
14-15	5.52	2.70	19.62	1.75	
15-16	5.98	3.35	16.93	1.62	
16-17	6.42	3.76	15.99	1.70	
17-18	6.84	3.67	15.60	1.69	
18-19	7.31	4.32	15.08	1.69	
19-20	7.74	3.53	14.85	1.68	
20-21	8.31	2.69	13.25	1.66	
21 - 22	8.70	2.00	12.45	1.65	
22-23	9.18	2.32	13.23	1.66	
23-24	9.66	1.76	11.65	1.64	
24 - 26	10.36	1.43	9.11	1.61	
26-28	11.10	0.71	7.97	1.60	
28-30	11.81	0.46	8.31	1.60	
30-32	12.81	0.0	8.89	1.61	
32-34	13.91	0.0	7.80	1.60	
34-36	14.91	0.0	8.64	1.61	
36-38	16.01	0.0	8.09	1.60	
38-40	17.01	0.0	5.14	1.57	
40-45	19.51	0.0	4.68	1.56	

<sup>\*</sup>The activity of supported  $^{210}\text{Pb}$  is  $3.0 \pm 1.5$  pCi g $^{-1}$ . The error in estimating excess  $^{210}\text{Pb}$  includes this uncertainty.

accounted for in terms of direct fallout to the water and instantaneous incorporation into sediments.

The most probable and self-consistent explanation for the lack of correspondence between atmospheric deposition and sedimentary records of the radionuclide is that a substantial portion is derived from the watershed. In large lakes such as the Great Lakes, very little <sup>137</sup>Cs originates from the watershed. In contrast, in a system such as a reservoir with a very short

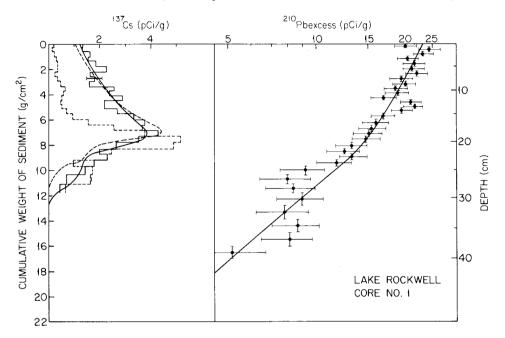


Fig. 6. Distribution of <sup>137</sup>Cs and excess <sup>210</sup>Pb in Lake Rockwell core 1. The solid curve in the <sup>210</sup>Pb plot is the distribution expected if the sedimentation rate is proportional to an exponentially increasing regional population and the activity of <sup>210</sup>Pb added to surface sediments (pCi g<sup>-1</sup>) is constant (CA model).

hydraulic residence time and a large (drainage basin)/(lake area) ratio, the watershed may contribute in a major way to the sedimentary load of the isotope. In contrast with  $^{137}\mathrm{Cs}$  reaching sediments from direct atmospheric transfer, that originating from the watershed will be integrated. That is, the load at a given moment will consist of a combination of all amounts deposited previously on the surrounding land. For purposes of illustrating the effects of watershed integration processes, it is assumed that the process may be characterized by a single time constant, the residence time,  $T_{\mathrm{D}}$ . If the flux of  $^{137}\mathrm{Cs}$  to the watershed is  $\Phi$  (pCi km $^{-2}$  yr. $^{-1}$ ), then the change in amount stored on the watershed L (pCi) is given by:

$$dL/dt = A_D \Phi - (\lambda + T_D^{-1})L \tag{1}$$

where  $A_{\rm D}$  is the area of the drainage basin; and  $\lambda$  is the radioactive decay constant for <sup>137</sup>Cs (= 0.69315/30.2 yr.<sup>-1</sup>). This equation has the solution:

$$L = \left[\exp\left(-\beta t\right)/\beta\right] \int_{0}^{t} A_{\mathrm{D}} \Phi \exp\left(\beta t\right) \mathrm{d}t \tag{2}$$

where  $\beta = \lambda + T_D^{-1}$ . The amount entering the reservoir is  $L/T_D$ . A computer program was used to evaluate eq. 2 and predict the distribution of <sup>137</sup>Cs in each core for different assumed residence times and sedimentation rates. The calculations are based on depths expressed in g cm<sup>-2</sup> to eliminate compaction effects. If  $g_k$  and  $g_{k+1}$  represent the cumulative weight of sediment at the bottom of the interval k and (k+1), respectively, then these values correspond to times (in years before 1977) of  $t_k = g_k/r$  and  $t_{k+1} = g_{k+1}/r$  where r is the sedimentation rate (g cm<sup>-2</sup> yr.<sup>-1</sup>). The expected activity in this time interval is then:

$$\overline{A}_k = \frac{\text{const.}}{t_{k+1} - t_k} \int_{t_k}^{t_{k+1}} L(t) \exp(-\lambda t) dt$$
(3)

The exponential term in the integral accounts for the radioactive decay of the isotope after deposition. The computer program finds a value of the constant which gives the best least-squares fit to the data for the choices of  $T_{\rm D}$  and r. Note that in reality the profile of <sup>137</sup>Cs is made up of contributions from both direct fallout and from the watershed. As the relative contributions cannot easily be distinguished on the basis of a few sedimentary profiles, the extreme case is initially chosen where the profile is considered to result only from integrated inputs.

The result of evaluating eqs. 3 and 4 for various choices of  $T_{\rm D}$  and r yields the smooth curves shown in Figs. 5 and 6. The least-squares optimized values of the parameters are given in Tables IV and V. Clearly a considerable improvement results if  $^{137}{\rm Cs}$  inputs are integrated either prior to or after their transfer to sediments. The theoretical curves exhibit an exponential decrease after the mid-1960's which reproduces the observed decline in  $^{137}{\rm Cs}$  activity. The fits are in fact of such excellent quality that no significant improvement results from adding any direct transfer component to the computation.

The single residence time integration concept is evidently applicable to these profiles, and it is likely that this integration is not localized in sediments to any significant degree. Benthic organism densities are insignificant and the presence of varves in the Mayfair Lake core argues against significant redistribution of sediment solids. Diffusional mobility cannot be ruled out on the basis of <sup>137</sup>Cs profiles alone, but the positions of the peak and horizon in the Lake Rockwell core 1 are correctly predicted from analysis of the <sup>210</sup>Pb profile below. The consistency of these very different approaches does rule out any major diffusional mobility of <sup>137</sup>Cs. Thus integrative effects must be non-local in character, but it cannot be determined if the integration occurs partly within the reservoir or is entirely a watershed effect. The

TABLE IV Mean linear sedimentation rates calculated by various methods

System	Mean date	Period	Mean sedimentation rate*1 (cm yr1)							
			stratigraphic	volumetric	radiom	population				
					<sup>137</sup> Cs	<sup>210</sup> Pb (CA)	<sup>210</sup> Pb (CF)	model*2		
Mayfair Lake	1980	1977—1982	2.4*3					2.4		
•	1967	1956-1977	1.3					1.5		
	1962	19381982	1.43	1.47				1.2		
	1949	19381956	0.83					0.93		
East Branch	1964	19501977			1.6			1.6		
Reservoir	1958	1939-1977		0.84				1.3		
	1945	1939-1950		0.90*4				0.90		
Lake Rockwell	1964	1950-1977			1.3*5	1.1	0.66	1.2		
	1958	1950-1965		1.0*6		0.86	0.56	0.89		
	1946	1914-1977		0.47		0.64	0.49	0.49		
	1932	1914-1950		0.27*4		0.20	0.38	0.29		

<sup>\*1</sup> Mean linear rates are obtained from the CA/population model.

\*2 Assumes the mean linear sedimentation rate is proportional to county population.

\*3 Linear sedimentation rates are not compaction-corrected but compaction generally amounts to a 10—15% effect.

<sup>\*4</sup> Hahn (1955) (dam closure to 1950).

\*5 Mean from two cores: No. 1: 1.4 cm yr.<sup>-1</sup>; No. 2: 1.2 cm yr.<sup>-1</sup>.

\*6 Hale & Kullgren Associates (unpublished data, 1982).

TABLE V System integration times based on constant and variable sedimentation rate models for the distribution of 137Cs

Core	Constant sedimer	ntation rate			Variable s	ediment	mentation rate			
	r (g cm <sup>-2</sup> yr. <sup>-1</sup> )	w(*1)	T <sub>D</sub> (yr.)	V(*2)	$A \simeq F(t)^{(*)}$	(3)	$A \simeq F(t)/r(t)^{(*4)}$			
	(g em yr. )	(em yr)	(y1.)		$T_{ m D}$ (yr.) $V$ $T_{ m D}$ (	T <sub>D</sub> (yr.)	V			
Lake Rockwell										
No. 1	0.57	1.4	8	0.34	6	0.51	11	0.46		
No. 2	0.55	1.2	20	0.24	15	0.32	<b>9</b> 5	0.29		
East Branch	0.85	1.6	10	0.38	7	0.43	14	0.38		

<sup>\*1</sup> Mean linear sedimentation rate.

\*2 
$$V = \left[ \sum_{i=1}^{N} (y_{\text{obs}} - y_{\text{calc}})^2 / (N-1) \right]^{\frac{1}{2}}$$

<sup>\*3</sup> CF analog model. \*4 CA analog model.

integration times resulting from application of the model are different in the two Lake Rockwell cores -8 yr. for core 1 and 20 yr. for core 2. This difference is real but its significance is uncertain. If the integration were solely a watershed process then values of  $T_{\rm D}$  should be indistinguishable from core to core. The observed trend toward increased mid-lake integration times could conceivably be related to time scales for horizontal redistribution of the isotope within the reservoir, but the data are too scarce to pursue this line of thought further.

# Profile of 210Pb

 $^{210}\text{Pb}$  is a naturally occurring radionuclide ( $t_{1/2}$  = 22.26 yr.) which has been used with increasing frequency to determine sediment accumulation rates in coastal marine and freshwater systems (Robbins, 1978). The isotope is produced in the atmosphere through radioactive decay of Rn gas and is efficiently scavenged from the air. In most freshwater environments it is strongly associated with suspended matter and rapidly transferred to sediments where its decay on burial provides the basis for a geochronology typically spanning about 100 yr. Its rate of delivery to water and land is essentially invariant from year to year, so the rate of transfer to a specific sedimentary site depends largely on its fate in the water column. In freshwater systems which have been affected by human activity (for example, through accelerated rates of eutrophication or altered loadings or eroded material), both the rate of transfer of <sup>210</sup>Pb and sediment as a whole may be changing in time although the rate at which the isotope is delivered to the water surface remains constant from year to year. A general model for the expected activity of <sup>210</sup>Pb in a continuously accumulating sediment column may be formulated by considering the rate of delivery of <sup>210</sup> Pb and sediment mass to be uncorrelated [further discussion of these ideas may be found in Robbins (1978)].

If r(t) is the rate of addition of sediment solids (in g cm<sup>-2</sup> yr.<sup>-1</sup>) and F(t) is the rate of transfer (flux) of excess <sup>210</sup>Pb (in pCi cm<sup>-2</sup> yr.<sup>-1</sup>), then the activity (pCi g<sup>-1</sup>) of material being deposited at the surface is:

$$A(t) = F(t)/r(t) \tag{4}$$

The flux, F, and activity, A, are expressed in terms of excess <sup>210</sup>Pb. Some <sup>210</sup>Pb is produced by the decay of Ra (or Rn) in the core itself. This supported activity must be subtracted from the total <sup>210</sup>Pb. In many cases the levels of supported <sup>210</sup>Pb are roughly an order of magnitude less than surface activities of total <sup>210</sup>Pb. The activity of excess <sup>210</sup>Pb at a depth in the core corresponding to a time t (yr.) before collection is then:

$$A(t) = F(t) \exp(-\lambda t)/r(t)$$
 (5)

where  $\lambda = 0.6932/22.26 = 0.03114 \text{ yr.}^{-1}$ .

When the sedimentation rate has changed over time as it apparently has in Lake Rockwell, there is no prior way to establish how F(t) has covaried with

r(t). It is customary to make certain extreme and simplifying assumptions about the nature of the covariance. In a system where increases in sedimentation rate are accompanied by corresponding increases in the sedimentary flux of <sup>210</sup>Pb, the ratio F(t)/r(t) at the time of deposition is constant and is equal to the activity of excess <sup>210</sup>Pb in surface sediments, A(0). This model is termed the constant activity (CA) or constant initial concentration (CIC, Appleby and Oldfield, 1978) model. Eq. 5 then reduces to:

$$A(t) = A(0) \exp(-\lambda t) \tag{6}$$

From this equation it follows that the age of a layer of sediment corresponding to depth Z (cm) and cumulative weight, g (g cm<sup>-2</sup>), will be given by:

$$t = -\lambda^{-1} \ln \left[ A(g)/A(0) \right] \tag{7}$$

This equation is termed the CA model age—depth relation. Sediment depths are expressed in terms of cumulative weight per unit area to eliminate small compaction effects.

In a system where the flux of <sup>210</sup>Pb remains constant while the rate of sediment accumulation changes, the age—depth relation may be established by integrating the activity of excess <sup>210</sup>Pb down the core. The total amount of excess <sup>210</sup>Pb in the core (in pCi cm<sup>-2</sup>) is

$$(\text{total}) = \int_{0}^{\infty} A(g) dg$$
 (8)

where dg is the thickness of each sediment interval (g cm<sup>-2</sup> yr.<sup>-1</sup>). This total activity must be equal to the total decay-corrected flux of <sup>210</sup>Pb, or:

$$\int_{0}^{\infty} A(g) dg = \int_{0}^{\infty} F \exp(-\lambda t) dt$$
 (9)

Similarly, the integrated activity to a depth g must correspond to the integrated decay-corrected flux:

$$\int_{0}^{g} A(g) dg = \int_{0}^{t} F \exp(-\lambda t) dt$$
 (10)

As F is assumed to be constant it may be eliminated from these equations to give the age—depth relation for the constant flux (CF) or constant rate of supply (CRS, Appleby and Oldfield, 1978) model:

$$t = -\lambda^{-1} \ln (1 - \zeta) \tag{11}$$

where

$$\zeta = \int_{0}^{g} A(g) dg / \int_{0}^{\infty} A(g) dg$$

[See Robbins (1978) for additional discussion.]

Both models represent conditions which in practice are only partially met. The CA model may be preferred for systems where the sources of increased sedimentation are exposed to inputs of excess lead <sup>210</sup>Pb (increased erosion of surface materials having appreciable excess lead <sup>210</sup>Pb activity) or increased scavenging of available <sup>210</sup>Pb from the water as a result of increased suspended-solids concentrations. The CF model may be expected to apply to systems where transfer of <sup>210</sup>Pb to sediments is already very efficient, so that additional inputs of particles cannot result in any significant additional deposition of the isotope. This would be the case, for example, in very large water bodies with long hydraulic retention times such as several of the Great Lakes where the residence time of <sup>210</sup>Pb is the order of a year. As the loading of excess <sup>210</sup>Pb to such lakes is almost entirely via direct atmospheric transfer, increases in sediment loads will not result in any significant increase in the flux of the isotope to sediments.

The distribution of excess  $^{210}$ Pb in Lake Rockwell core 1 is shown in Fig. 6. If both the sedimentation rate and flux were constant, the log of the activity would decrease linearly within sediment depth. The observed profile exhibits a striking curvature which cannot be ascribed to either analytical or stratigraphic factors. The gross sediment properties (color, texture) as well as the fraction of sediment soluble on prolonged acid extraction show no significant change with depth while the bulk density, fraction dry weight and porosity increase gradually with depth as a result of compaction but exhibit no discontinuities. The error bars associated with the excess activities reflect combined errors in activities of total and supported  $^{210}$ Pb. The activity of  $^{210}$ Pb at the surface is  $25.9 \pm 1.0$  pCi g<sup>-1</sup> while the activity of supported  $^{210}$ Pb is  $3.0 \pm 1.5$  pCi g<sup>-1</sup>.

The age—depth relations are provided in Fig. 7 for the two alternative models. Uncertainties in the determination of A(0) and the supported <sup>210</sup>Pb activity have been propagated through the calculation of the age of each section in the case of the CA model to illustrate the level of uncertainty in age determinations and its increase with sediment depth. The CA and CF models give very different predictions of the age of sediment sections in this core. The contrast between model predictions increases with sediment depth, with the CF model predicting a lower rate of sedimentation than the CA model. It has been shown that this is a general difference between the two models when sedimentation rates are increasing (Robbins, 1978). Note that in the case of the CA model, some estimates of sediment ages near the surface are negative due to random errors in the activities. Because of the way in which the age—depth relation is established for the CF model, negative ages can never be obtained from that relation. The solid line through each set of data points is a portion of a parabola with coefficients determined via least-squares optimization.

Agreement between the age—depth relationships and a second-order polynomial indicates that the excess <sup>210</sup>Pb profile is consistent with a linearly increasing sedimentation rate. However, the age—depth relation based on the

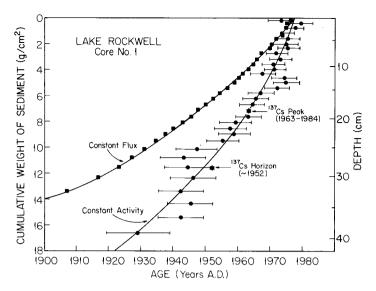


Fig. 7. The relation between sediment depth (in g cm<sup>-2</sup>) and model age for the constant activity (CA) and constant flux (CF) model in Lake Rockwell core 1. The stars indicate the position of the <sup>137</sup>Cs peak (1963–1964) and horizon (1952). The <sup>137</sup>Cs data as well as other data support the choice of the CA model for these systems. The CF model systematically underestimates the observed rate of sediment accumulation.

CA model is also consistent with county population increases since about 1900. The time dependence of the population since 1900 is well approximated by the expression:

$$P(t) = P_0 + P_1 \exp(-\beta t) \tag{12}$$

where P(t) is in thousands; and t is the time before 1977. Least-squares values for the coefficients are,  $P_0 = 15.3$ ,  $P_1 = 67.7$ , and  $\beta = 0.0635$  yr.<sup>-1</sup>. The sedimentation rate is then given by:

$$r(t) = \alpha P(t) \tag{13}$$

and the age—depth relation by:

$$g(t) = \int_{0}^{t} r(t)dt = \alpha [P_0 + (P_1/\beta)\{1 - \exp(-\beta t)\}]$$
 (14)

where the least-squares value of  $\alpha$  for the CA age—depth relation is 0.00896. The resulting curve is indistinguishable from that shown in Fig. 7 for a parabolic fit. With the CF model, the relation between the age and depth is poorly reproduced by the assumption that r(t) is proportional to P(t). The theoretical excess <sup>210</sup>Pb profile shown as the smooth curve in Fig. 6 is based on the CA model and the above relation (eqs. 12 and 13) between sedimenta-

tion rate and population size. The resulting sedimentation rate and county population data are shown in Fig. 8.

While the correspondence between sedimentation rate and regional population is significantly better using the CA model, that is only a weak argument for preferring it over the CF model for this lake. However, there are several more compelling arguments. First, the distribution of <sup>137</sup>Cs is more consistent with the CA model. The position of the peak (1963—1964) and the horizon (1952) are shown on the age—depth plot (Fig. 7). Note that both points fall very close to the relation obtained from the CA model but are completely inconsistent with the CF model curve. Furthermore, the CA model time dependence of the sedimentation rate is consistent with the volumetric measures of sedimentation rate whereas the CF model rates are not.

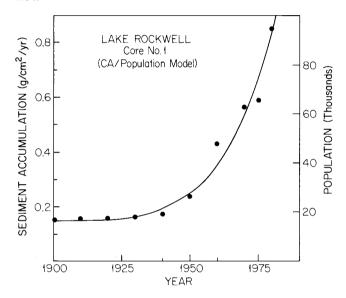


Fig. 8. The mass sedimentation rate (g cm<sup>-2</sup> yr.<sup>-1</sup>) in Lake Rockwell (core 1) vs. year based on an exponential population growth model (eq. 13). Individual points are county population census data. There is no a priori reason why future increases in population should be accompanied by corresponding increases in sedimentation rate.

### Comparison of sedimentation rates

The mean sedimentation rates for various periods and methods for the three systems are summarized in Table IV. Because some observations are reported in linear units, all values have been converted to mean linear sedimentation rates for purposes of comparison. For Lake Rockwell the volumetrically determined sedimentation rates agree well with the CA/population model values for <sup>210</sup>Pb and with rates based on analysis of <sup>137</sup>Cs profiles in two cores. As expected, the CF model values shown for comparison do

not agree as well. Since volumetrically determined sedimentation rates represent an average over the entire lake, it is clear that the two cores are representative of the lake as a whole. The increase in sedimentation rates over the past few decades is also seen in the varve data from Mayfair Lake and to a less compelling extent in the East Branch Reservoir core as well. For comparison, the population model has been evaluated to provide a rough prediction of the mean linear sedimentation rate for each period. Increases in sedimentation rate in all three reservoirs are consistent with increases in regional population since 1900.

It is likely that the long-term increase in sedimentation rate in the three reservoirs is due to the increasing human use — primarily residential, commercial and light manufacturing — of the drainage basins. Most of the drainage basins of Lake Rockwell and East Branch is in Geauga County, Ohio. The population increase in the county dates from the 1930's, which is also the time over which the sedimentation rate in Lake Rockwell has been increasing. The doubling time of population size is ~19 yr., a value seen to be very similar to the doubling time of sedimentation rate in all three reservoirs. It is likely that most of the increased load of sediment delivered to the reservoir is due to increased erosion and not to increased runoff. Annual averages of runoff from Lake Rockwell are variable ( $\bar{x} = 7.93 \text{ m}^3 \text{ s}^{-1}$ ; s = 1.78), but a linear regression on the flow data indicates that there has been only a 26% increase in Lake Rockwell outflow (6.74—8.50 m³ s<sup>-1</sup>) from 1940 to 1980. The increase in sedimentation rate over the same time interval is over 10 times larger (381%). The higher sedimentation rate in Mayfair

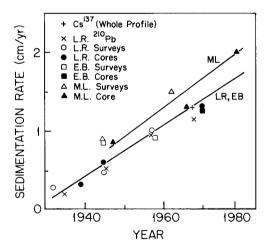


Fig. 9. The mean linear sedimentation rate vs. year for all three systems based on various chronological methods. In each reservoir sedimentation rates have more than tripled since about 1940. Sedimentation rates and the increase in time are indistinguishable in East Branch Reservoir and Lake Rockwell. Rates are  $\sim 20\%$  higher in Mayfair Lake.

Lake may be due either to the difference in the ratio of drainage area to basin volume or to the fact that the population density in the drainage basin of Mayfair Lake has greatly increased since the 1950's and is now  $\sim 10$  times that of Lake Rockwell (Garner, 1982).

The changes in mean linear sedimentation rate in the three reservoirs as determined by the several different methods are summarized in Fig. 9.

## <sup>137</sup>Cs and variable sedimentation rates

In the above discussion of <sup>137</sup>Cs profiles, theoretical distributions were calculated assuming that over the period from about 1950 to 1977, the sedimentation rate was constant. If they have increased considerably over the period, as several lines of evidence suggest, then theoretical profiles should be recomputed using the best estimates of r(t) and some assumption about the covariance of the loading of <sup>137</sup>Cs to the system as a function of r(t). Although the evidence suggests that for <sup>210</sup>Pb, the flux and r(t) are well correlated, it is not clear how to specify this covariance under non-steadystate conditions for <sup>137</sup>Cs. Two contrasting models offer themselves in the absence of specific knowledge of how changing rates of erosion of materials from the drainage basin affect the transfer of 137Cs. Both models assume that the loading of the radionuclide to the reservoir is given by eq. 2, so that the storage on the drainage basin is characterized by a constant residence time. The first model, in analogy with the constant activity model, assumes that the activity at the time of deposition is proportional to this loading so that the activity with depth is:

$$A(g) = (\text{const.}) L(t) \exp(-\lambda t)$$
 (15)

where the age—depth relation (g vs. t) is given by the CA/population model from <sup>210</sup>Pb. The above relation envisions that the increase in loading of the isotope is accompanied by a corresponding increase in sedimentation rate. The alternative model assumes that the loading of <sup>137</sup>Cs is completely uncorrelated with sedimentation rate so that the profile is given by:

$$A(g) = (\text{const.}) L(t)/r(t) \exp(-\lambda t)$$
(16)

This model is analogous to the CF model. In the case of <sup>137</sup>Cs it is clear that neither model is strictly correct since increases in sedimentation rate must be accompanied by a decrease in watershed residence time. However, that relationship is not specifiable a priori and cannot be reliably inferred from analysis of <sup>137</sup>Cs distributions in only a few cores and little supplementary information about characteristics of the drainage basin. Thus the two models serve primarily to illustrate the effects of variable sedimentation rates on such distributions but have limited usefulness in characterizing the nature of the integrative processes. The results of applying the CA analog model are shown as the smooth (dashed line) curves in Figs. 5 and 6. The general result

of introducing variable sedimentation rates and the CA analog model is to: (1) reduce the apparent watershed residence time; (2) sharpen the peak; and (3) reduce the apparent range of penetration of  $^{137}$ Cs near the horizon. The quality of the fits is uniformly worse, but not by much. Theoretical distributions of  $^{137}$ Cs using the CF analog model (eq. 16) are not significantly different from the CA model distributions shown and are in fact experimentally indistinguishable. The major difference between the two models is in the estimates of system integration time. As shown in Table V, the CF analog model predicts longer integration times than the CA model. This arises because division of the loading (L in eq. 16 above) by a sedimentation rate which is increasing toward the sediment surface results in a relatively lower predicted activity which must be compensated by increased integration in order to reproduce the observed distributions.

While both variable sedimentation rate models improve the location of the peak position, they underestimate the observed penetration of the isotope near the horizon by  $\sim 1$  cm (0.5 g cm $^{-2}$ ). The discrepancy can be removed by assuming a higher sedimentation rate in the 1950's than that based on  $^{210}$ Pb (constant sedimentation rate model) or alternatively by invoking some slight diffusional mobility of the isotope. An effective diffusion coefficient associated with the spread in a concentration front is given by Crank (1957):

$$D_{\rm e} \simeq \Delta Z^2 / 2\Delta t \tag{17}$$

where  $\Delta Z$  is the characteristic length; and  $\Delta t$  is the elapsed time. In this core  $\Delta Z$  is the order of 1 cm/20 yr., so from eq. 17 the effective diffusion coefficient would be around 0.03 cm<sup>2</sup> yr.<sup>-1</sup>. The effective molecular diffusion coefficient is given by Li and Gregory (1974):

$$D_{\mathbf{e}} \approx \alpha \theta^{-2} (1 + K)^{-1} D_0$$

where the term  $\alpha\theta^{-2}$ , is nearly unity for these sediments; K is the dimensionless distribution coefficient; and  $D_0$  is the diffusion coefficient of  $^{137}\mathrm{Cs}$  in free solution ( $\sim 150~\mathrm{cm^2}~\mathrm{yr.^{-1}}$ ). The above values imply a distribution coefficient of  $5 \cdot 10^3$  which is comparable to values reported by others for short-term uptake experiments with freshwater sediments (cf. Robbins et al., 1979). While some limited mobility of the isotope appears plausible, it should be noted that the mobility of  $^{137}\mathrm{Cs}$  depends strongly on the type of sediments in question and that the distribution coefficient for the Lake Rockwell sediments is unknown.

The effect of varying system integration times on the distribution of  $^{137}$ Cs is illustrated in Fig. 10. The calculations are based on the CA analog model (eq. 15). Note that for integration times comparable to the half-life of  $^{137}$ Cs, the calculated distribution varies considerably near the interface and at the peak. However, for times greater than  $\sim 50$  yr., calculated profiles are very insensitive to the assumed value of  $T_r$ . In fact  $T_r = 50$  yr. is experimentally

indistinguishable from  $T_{\rm r}$  = 5000 yr.! Varying the integration time has essentially no effect on the location of the <sup>137</sup>Cs horizon, since the integration occurs outside the sediment core.

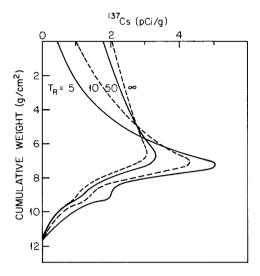


Fig. 10. Theoretical distributions of  $^{137}$ Cs in the Lake Rockwell core I for various system integration times. Near-surface and peak activities are especially sensitive to changes in the integration time between 0 and  $\sim 30$  yr. For times greater than 50 yr., profiles are extremely insensitive to the choice of time.

# Total 137Cs and excess 210Pb

The total amount of <sup>137</sup>Cs and <sup>210</sup>Pb in each core, provided in Table VI, may be compared with the amount expected via direct atmospheric transfer. The average amount of <sup>137</sup>Cs in the two Lake Rockwell cores is 22.5 pCi cm<sup>-2</sup> as compared with an atmospheric load of ~8 pCi cm<sup>-2</sup> [decay corrected to 1977 (see Edgington and Kartunnen, 1977)]. In Lake Rockwell core 1 the total excess <sup>210</sup>Pb is 231 pCi cm<sup>-2</sup> as compared with an expected standing crop of 23 pCi cm<sup>-2</sup> based on regional atmospheric deposition measurements (Talbot and Andren, 1983). Thus ~3 times as much <sup>137</sup>Cs and 10 times as much 210Pb has accumulated at these locations as can be accounted for by direct atmospheric deposition. For 137Cs this factor is comparable to that found in other reservoir/drainage basin systems. Ritchie et al. (1973) reported that the concentrations of <sup>137</sup>Cs per unit area of reservoir sediments in three small watersheds in northern Mississippi were 2.8, 3.8 and 4.0 times that of the soils in their respective watersheds, thus indicating that the reservoirs were acting as traps for this radionuclide. There are two alternative explanations for the enhanced deposition: either these radionuclides are focused into selected areas of the lake or the drainage basin is a major source. Without far more extensive coring of the lake itself it is impossible to rule out focusing. However, the two cores are apparently

TABLE VI

Cumulative deposition of <sup>137</sup>Cs and excess <sup>210</sup>Pb in sediments compared with atmospheric deposition

	Total activity (pCi cm <sup>-2</sup> )		
	<sup>137</sup> Cs	<sup>210</sup> Pb	
Lake Rockwell			
No. 1	25.5	231	
No. 2	19.5		
East Branch	39.9	_	
Atmospheric	8*1	23*2	

<sup>\*1</sup> Edgington and Kartunnen (1977).

representative of average sedimentation in the lake and do not differ greatly in their total <sup>137</sup>Cs content. Thus drainage basin sources appear to be important. If the lake were a perfect trap for these radionuclides then the single residence time model above would provide an estimate for the average activity in the lake bottom. In eq. 1 the rate at which <sup>137</sup>Cs is lost from the drainage basin can be seen to be  $(\lambda + T_D^{-1})L(t)$  in pCi yr. The first term,  $\lambda L(t)$ , is the loss via radioactive decay while the second term,  $T_D^{-1}L(t)$ , is the loss via erosion and runoff. If  $A_L$  is the area of the lake, then the amount (pCi cm<sup>-2</sup>) in sediments due to runoff is:

$$(\text{total}^{137}\text{Cs}) = A_{\text{L}}^{-1} \int_{0}^{t} T_{\text{D}}^{-1} L(t) \exp(-\lambda t) dt$$
 (19)

provided no 137Cs is lost.

However, since the hydraulic residence time for Lake Rockwell is only  $\sim 9$  days, the system is obviously not a very good trap for water and thus losses of particle-associated tracers could be very large as well. To account for the imperfect trapping of particles in the system, we introduce a particle residence time for the lake,  $T_{\rm L}$ . The total amount of <sup>137</sup>Cs in the lake—sediment system, S (pCi), then is given by:

$$dS/dt = A_{L}\Phi + T_{D}^{-1}L - (\lambda + T_{L}^{-1})S$$
(20)

where the first term,  $A_{\rm L}\Phi$ , is the loading directly from the air (pCi yr.  $^{-1}$ ). In this equation and eq. 1, the unknowns are  $T_{\rm D}$  and  $T_{\rm L}$ . Assuming that both  ${\rm d}L/{\rm d}t$  and  ${\rm d}S/{\rm d}T=0$  for  $^{210}{\rm Pb}$  eqs. 1 and 20 reduce to:

$$L = A_{D}\Phi/(\lambda + T_{D}^{-1})$$

$$S = \Phi[A_{L} + A_{D}(1 + \lambda T_{D})^{-1}]/(\lambda + T_{L}^{-1})$$
(21)

<sup>\*2</sup> For Crystal Lake, northern Wisconsin (Talbot and Andren, 1984).

The total activity of  $^{210}$ Pb per unit area of the lake is then  $S/A_{\rm L}$  and hence:

$$P (pCi cm^{-2}) \equiv S/A_{L} = \Phi [1 + A_{D}A_{L}^{-1}(1 + \lambda T_{D})^{-1}]/(\lambda + T_{L}^{-1})$$
 (22)

and the relation between  $T_{\rm L}$  and  $T_{\rm D}$  is:

$$T_{L} = \gamma / [\lambda \{1 - \gamma + A_{D}A_{L}^{-1}(1 + \lambda T_{D})^{-1}\}]$$
(23)

and  $\gamma$  is the ratio of the total excess <sup>210</sup>Pb to the standing crop,  $\gamma = P(\Phi/\lambda)^{-1}$  = 10 (see Table VI). For values of the system integration time implied by distributions of <sup>137</sup>Cs, ~10–20 yr., the value of  $T_{\rm L}$  is ~2 yr. as determined from the above equation. Note that as  $T_{\rm D}$  increases, the loading of <sup>210</sup>Pb from the drainage basin decreases, so for a given value of  $\gamma$ ,  $T_{\rm L}$  must increase to compensate for reduced inputs. For complete trapping of particles, in the reservoir ( $T_{\rm L} = \infty$ ), the system integration time,  $T_{\rm D}$ , is ~650 yr. Thus integration times greater than this value cannot produce the observed deposition of <sup>210</sup>Pb.

The non-steady-state solution of eqs. 1 and 20 for  $^{137}\text{Cs}$  gives a value of  $T_{\rm L}$  of  $\sim 0.5$  yr. for values of  $T_{\rm D}$  in the range of 10–20 yr. As the models are only approximate and the atmospheric loadings are poorly known, it is not clear if the difference between values of  $T_{\rm L}$  based on  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  are real. For the East Branch Reservoir a calculation gives a residence time of  $\sim 5$  yr. for a watershed residence time of 10–20 yr. These residence times correspond to 13% of the  $^{137}\text{Cs}$  and 60% of the  $^{210}\text{Pb}$  retained in Lake Rockwell per year and to 82% of the  $^{137}\text{Cs}$  retained in East Branch Reservoir per year. In the study of three reservoirs by Ritchie et al. (1973), the trapping efficiencies were 57%, 38% and 25%, respectively. Thus the values obtained in this analysis are consistent with previously reported values.

In Lake Rockwell, losses predicted from a watershed erosion model are also in general agreement with activities observed in sediment cores. Ritchie et al. (1973) found that, in general, the total amount of <sup>137</sup>Cs lost from the watershed could be related to the erosion rate (t ha<sup>-1</sup> vr.<sup>-1</sup>)\* by:

$$(\% \text{ of total}) = 1.6 \times (\text{erosion rate})^{0.68}$$
(24)

The erosion rate inferred from the data in Table I, assuming a mean density of sediment solids of 2.6 g cm<sup>-3</sup>, is 0.64 t ha<sup>-1</sup> yr.<sup>-1</sup>. From the above relation the expected loss of the radionuclide would be 1.2% of the total stored on the watershed or 0.51 Ci. The amount contributed to sediments is then ~18 pCi cm<sup>-2</sup> plus another 8 pCi cm<sup>-2</sup> from direct atmospheric transfer for a total of 26 pCi cm<sup>-2</sup>. The observed activity is 23 pCi cm<sup>-2</sup>. For the East Branch Reservoir the activity is about twice as high as that predicted by the above relation (see Table VII). Considering the approximations involved, the agreement is satisfactory.

<sup>\*1</sup> t = 1 tonne =  $10^3$  kg; 1 ha = 1 hectare = 0.01 km<sup>2</sup>.

TABLE VII

Expected sediment inventories of <sup>137</sup>Cs based on a semi-empirical model of loss vs. watershed erosion rates\*<sup>1</sup>

	Lake Rockwell	East Branch Reservoir	Mayfair Lake
$A_{\rm D} (10^6 {\rm m}^2)$	531	45.3	2.4
Total <sup>137</sup> Cs (Ci)	42.5	3.62	0.19
Erosion rate*2	0.64	8.15	2.47
Per cent lost*3	1.2	6.7	3.0
<sup>137</sup> Cs lost (Ci)	0.51	0.24	0.0057
$A_{\rm L} (10^6 {\rm m}^2)$	2.78	1.70	0.0155
Mean activity from watershed i	nput		
(pCi cm <sup>-2</sup> )	18	14	36
Atmospheric input			
(pCi cm <sup>-2</sup> )	8	8	8
Total expected*4			
(pCi cm <sup>-2</sup> )	26	22	45
Observed (pCi cm <sup>-2</sup> )	23	40	_

<sup>\*1</sup> Ritchie et al. (1973).

# Utility of the sedimentation rate calculations

Rivers and streams are often impounded for recreational purposes and to create surface water supplies. As soon as a dam is closed, the reservoir begins to fill with river-borne sediment. The sediments themselves can be considered to be pollutants, in the sense that they shorten the useful life of the reservoir, necessitating expensive remedial measures (F.W.Q.A., 1970; E.S.W.G., 1975). The volume of a reservoir basin divided by the rate of annual accumulation of sediment gives an estimate of the time to complete infilling of the basin. However, at some point before it is completely filled, an impoundment will become useless because of turbidity, growth of algae and rooted aquatic vegetation, or large water-level fluctuations. Stevens (1936) took this point to be 75% of total capacity; Hahn (1955) estimated it to be 60% of total capacity.

Both Lake Rockwell and East Branch Reservoir are only  $\sim 10\%$  full at present. Assuming that the long-term sedimentation rate does not change, Lake Rockwell has 203 more years of useful life. Of course, the sedimentation rate has not been constant, and the present rate is  $\sim 3$  times greater than the long-term average (Table I; Fig. 3). If the present sedimentation rate remains unaltered in the future, there is only about 67 more years of useful life remaining for water-supply reservoir.

<sup>\*2</sup> Assumes a mean density of sediment solids of 2.6 g cm<sup>-3</sup>.

<sup>\*3</sup> Per cent of  $^{137}$ Cs lost = 1.6 × (erosion rate) $^{0.68}$ .

<sup>\*4</sup> Assumes 100% retention of <sup>137</sup>Cs in the reservoir.

Mayfair Lake, on the other hand, is already  $\sim 50\%$  full of sediment, and with an unchanged long-term rate of infilling, will have only seven more years of useful life. In fact, the sediment survey of Mayfair Lake was prompted by complaints from shoreline residents of algal growth, foul odors in the lake, and shallowing of the upstream portion of the reservoir. While some improper sewer connections upstream of the lake have exacerbated pollution problems (Garner, 1982), such problems are typical of aging lakes. If the present high rate of sedimentation continues, Mayfair Lake will have less than five more years of useful life.

Knowledge of sedimentation rates is also essential for calculating the flux of pollutants associated with sediment particles. Hydroxide coatings on clay particles bind algal nutrients such as phosphorus and toxic materials such as heavy metals, pesticides and PCB's. Bottom sediments are not only a sink for hazardous materials, but also a short-term source. Materials can be remobilized in at least two ways. Sediment particles may be resuspended under the influence of waves and currents, or particles may be buried and eventually pass through different chemical regimes where their coatings dissolve. Dissolved solutes may then diffuse or be advected back to the sediment—water interface.

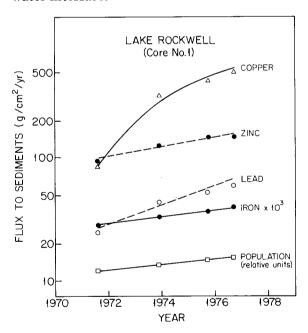


Fig. 11. Metal fluxes since 1970 in Lake Rockwell core 1. The flux of Fe, whose concentration has not changed significantly, increases in proportion to population size corresponding to increases in sedimentation rate. Fluxes of Pb and Cu increase more rapidly because of additional loadings to the system. CuSO<sub>4</sub> was added to the lake in large amounts during the 1970's as an algicide.

Trace-metal concentrations within the top 12 cm (7 yr.) of sediment were measured in Lake Rockwell and East Branch Reservoir. In Lake Rockwell, metal concentrations decreased with depth in the core (Table VIII), which indicates that the mass flux of metals to the bottom has increased with time along with the sedimentation rate. Within East Branch Reservoir, concentrations of all metals were more or less constant with depth in the core. Depositional fluxes of metals can be calculated as the product of metal concentration (mg/g dry-weight sediment) from Table VIII and the sedimentation rate (g dry-weight sediment cm<sup>-2</sup> yr.<sup>-1</sup>) from Fig. 8. Fig. 11 shows the results of the flux calculations. In East Branch Reservoir the tracemetal concentrations are essentially constant. Any increases in metal deposition rates are associated with increased rates of sedimentation. Similarly, in Lake Rockwell Fe and Zn concentrations show 10% and 20% increases, respectively, between 1970 and 1977. During the same period the sedimentation rate has increased by  $\sim 30\%$ , so the increase in flux is due to both increased concentrations and rates of sedimentation. Because increasing sedimentation is a major factor governing the time dependence of fluxes of these metals, their increase parallels the increase in regional population during this period (see Fig. 11). In contrast, there is a nearly two-fold increase in the concentration of Pb, and only 30% of the change in flux is due to increased sedimentation. Therefore, most of the increase is attributable to increases in the rate of addition of the metal to the system presumably as a

TABLE VIII

Total metal content of sediment core sections from East Branch Reservoir and Lake Rockwell\*

	Zn (ppm)	Cu (ppm)	Pb (ppm)	Fe (ppt)
East Branch				
0—2 cm	129	31	43	42.0
2-4 cm	139	33	45	41.3
5—7 <b>cm</b>	140	32	36	41.3
7—12 cm	138	40	29	44.7
Lake Rockwell				
0—2 cm	205	673	84	56.4
2-4 cm	214	620	76	53.1
5—7 <b>cm</b>	198	505	68	53.7
7—12 cm	167	147	44	51.3
Cuyahoga River				
Mouth (averages, 1970)	1,236	169	565	75.8
U.S. Environmental Protection Agency region IX dredged material				
disposal criteria	75	50	50	_

<sup>\*</sup>Transect No. 3.

result of the continuing widespread use of leaded fuel additives and the subsequent deposition of combustion products on the watershed and directly into the reservoir.

Particularly dramatic is the increase in deposition rates of Cu from 1970 to 1977. At the present time, Lake Rockwell surface sediments receive ~10 times the amount of Cu per year as sediments of East Branch Reservoir. Concentrations of Cu are 10 times those recommended for "clean" sediments (Table VIII). Most of the Cu is probably the result of the addition of CuSO<sub>4</sub> to the reservoir to control algal growth in the water column in late summer. Historical records show that  $\sim 2.34 \cdot 10^7$  g Cu were added to the reservoir over the period 1970–1977. Based on a single core,  $\sim 4.2 \cdot 10^7$  g Cu are stored in the lake sediments for the same period. Thus a considerable fraction of the sediment inventory is apparently attributable to direct additions of Cu to the lake. The effects of these additions on fish or bottom fauna are unknown, but bacterial populations have apparently been little affected. The standing crop of bacteria in Lake Rockwell is over twice that in East Branch Reservoir  $(11.7 \cdot 10^9 \text{ vs. } 4.7 \cdot 10^9 \text{ bacteria/g dry-weight sediment}).$ Some of the differences may be due to the sulphate, a bacterial nutrient, added with the Cu. Bacteria remineralize organic matter, which raises the possibility that a material added to prevent algal growth may over the longerterm result in increased production.

#### CONCLUSIONS

In this paper we have shown that several different approaches to determining sedimentation rates in reservoirs are generally self-consistent and indicate that rates of sediment accumulation have increased dramatically since the mid 1940's in three separate systems. Within experimental uncertainties, sedimentation rates expressed in centimeters per year have increased linearly since about 1940 and mass sedimentation rates are essentially proportional to exponential increases in regional population during the past three decades.

Mayfair Lake has a higher sedimentation rate than Lake Rockwell and a 10 times higher population density. The association of increased population size with increased reservoir sedimentation rate and trace-metal flux is typical of a number of other different reservoirs with a variety of soil types (Guy and Ferguson, 1962; Kautzman and Cavaroc, 1973). A 25% increase in average runoff in Lake Rockwell from 1941 to 1978 may also be due to urbanization. Because of increased rates of sedimentation, the useful life of Lake Rockwell has already been reduced from 203 to 67 yr. If sedimentation rates continue to increase with population growth, the useful life of this and related systems in the region could be even more strikingly diminished.

As these reservoirs possess a well-established change of sedimentation rate, they provide a means for testing alternative models for <sup>210</sup>Pb dating. The constant activity (CA) model is clearly preferred over the constant flux

model (CF) in these systems. In the CA model the flux of excess <sup>210</sup>Pb is proportional to the rate of sediment accumulation. Evidently increases in the rate of erosion from the drainage basin results in the introduction of materials to the reservoir which have appreciable excess <sup>210</sup>Pb at concentrations which have not changed greatly with urbanization of the drainage basin. <sup>210</sup>Pb is evidently a useful tool for determining changing rates of sedimentation in reservoirs, but inferred sedimentation rates depend critically on the models used in analysis of profiles. The validity of the <sup>210</sup>Pb method applied to systems with changing sedimentation rates should always be checked by alternative geochronological techniques.

Distribution of fallout <sup>137</sup>Cs exhibits far higher surface activity than expected from direct atmospheric deposition. This feature is the result of integrative processes occurring within the reservoir—watershed system. The integration process, characterized by times of one to several decades, is probably not localized within sediments but is either the result of sediment redistribution processes within the reservoir or, more likely, a property of the drainage basin itself. Roughly 3 times as much <sup>137</sup>Cs and 10 times as much <sup>210</sup>Pb are deposited in sediments than can be due to direct transfer from the atmosphere. If the cores are representative of each reservoir, then drainage basins are contributing most of these radionuclides to the reservoirs. For drainage basin residence times on the order of 10—20 yr. the reservoirs must be between 14% and 82% efficient in trapping <sup>137</sup>Cs and 60% efficient in trapping <sup>210</sup>Pb to produce the observed total amount in sediments.

### ACKNOWLEDGEMENTS

The authors thank K.A. Johansen of the University of Michigan, Great Lakes Division, for his assistance in preparation and counting some of the sediment samples. Kim McCullough and Sharon Matis assisted in the field and made laboratory geochemical determinations. Charles Hahn (O.D.N.R.) provided his field notes and useful conversation about his earlier surveys on these reservoirs. They thank also the City of Akron Department of Public Service, Public Utilities Bureau-Water, and especially Mr. Joseph Hobroken, for assistance in the field and for information about the water-supply reservoirs. This work was supported by grants to P. McCall from the Northeast Ohio Areawide Coordinating Agency and to G. Matisoff from the Cuyahoga County Regional Sewer District. This study was partially carried out while one author (J.A.R.) was at the University of Michigan, Great Lakes Research Support of that organization is gratefully acknowledged. Division. Contribution No. 373 of the Great Lakes Environmental Research Laboratory.

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